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Materials and Structures

Applied Ceramic Research Program

SEMIANNUAL TECHNICAL REPORT

(1 January–30 June 1962)

12 OCTOBER 1962

Prepared by E. G. KENDALL
Materials Sciences Laboratory

Prepared for DEPUTY COMMANDER AEROSPACE SYSTEMS
AIR FORCE SYSTEMS COMMAND
UNITED STATES AIR FORCE
Inglewood, California

LABORATORIES DIVISION • AEROSPACE CORPORATION

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ABSTRACT

The major construction phase for establishing a suitable laboratory for ceramic research has been completed and the procurement of many key equipment items for this research has been concluded. Descriptions of key pieces of equipment that give the Aerospace Ceramic Laboratory a unique capability are presented.

Although construction of the laboratory has seriously hampered significant research progress, work of a qualitative exploratory nature has been performed and is reviewed.

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I. INTRODUCTION

An urgent requirement exists for the development of high-temperature resistant materials for structural and propulsion systems applications. Materials possessing adequate mechanical and physical properties are required for use at temperatures from $2000^{o}F$ to over $5000^{o}F$ for special applications in future Air Force systems. The ceramic materials possessing these qualities include primarily refractory compounds in the forms of carbides, borides, nitrides, oxides, and silicides.

Despite an increased interest in ceramic materials in recent years, the potential of these materials for aerospace use has not been determined. In most cases this is a reflection of the failure to obtain information on how microstructure and purity affect properties. Since both microstructure and purity are controlled by processing techniques, the objective of this program is to improve the mechanical and physical properties of ceramic materials by acquiring a basic scientific understanding of processing variables and their effect on the important properties for aerospace systems.

II. DISCUSSION

The general background for the Applied Ceramic Research Program has been given in two previous reports[1,2].

A. LABORATORY FACILITIES

During this six-month period, the major construction phase for establishing a suitable ceramic laboratory has been completed; also, the procurement of many key equipment items for this research has been concluded. It should be noted that this equipment, in many cases, has been especially designed by Aerospace personnel to provide unique and varied capabilities for the study of ceramic materials.

Descriptions of some of the more important equipment for detailed studies on ceramic systems are listed as follows:

1) A non-consumable electrode arc furnace (Figure 1), especially designed by Aerospace personnel, has been placed in operation. This furnace, powered by DC rectifiers, operates efficiently at 3000 amps and 30 volts for both alloying and casting refractory materials. Standard water-cooled copper or graphite crucibles and molds may be utilized in this furnace. Material applications to date include columbium, tungsten, titanium, titanium carbide, and boron.

2) A high-temperature (2500°C) resistance-heated vacuum furnace (Figure 2) has been acquired. The tantalum heating element is a square configuration that yields a 4-inch by 4-inch hot zone 7 inches in length. The vacuum system is capable of evacuating the furnace chamber to levels of 1×10^{-5} Torr.

3) An apparatus for the hot pressing of refractory powders in an argon atmosphere, designed and fabricated by Aerospace personnel, is shown in Figure 3. This hot press is horizontally loaded and induction heated with the hydraulic pressure system being capable of generating a 10,000-psi force across a 4-inch diameter specimen face. The induction system, operated at 3600 cps with a rating at 30 kw, is capable of heating a graphite die and susceptor assembly to 2000°C.

4) An arc melting skull-casting vacuum furnace which can melt by either consumable or non-consumable electrodes has been acquired and is shown in Figure 4. The furnace is equipped with 5- and 10-pound skulls, power requirements which can allow melting at 5000 amps and 30 volts, and a 230 cfm mechanical booster vacuum pump for system evacuation. The tilt—pour crucible and power system enables vacuum melting and subsequent casting directly to specimen size for refractory materials with melting points as high as 4000°C.

5) A hydrogen-atmosphere muffle furnace with a molybdenum heating element has been designed and is being fabricated. This furnace will be capable of continuous operation at 1700°C with a usable hot zone 28 inches long. The D-shaped alumina muffle has a 4-inch by 6-inch cross section and supports the molybdenum heating element. Overall length of the furnace is 10-1/2 feet, including a 36-inch long water-cooled section. End purge chambers, automatic flame seals, and a hydrogen drier all ensure the maintenance of a low dewpoint within the furnace to allow sintering of refractory metals, compounds, and ceramics.

6) An isostatic press (Figure 5) has been designed and fabricated. This press has a high-pressure chamber 6 inches in diameter and 12 inches long. The hydraulic system is capable of exerting pressures of 20, 000 psi at room temperature.

7) An electron beam zone-refining and crystal-growth apparatus has been designed and is being fabricated. The vacuum chamber consists of a stainless steel bell jar with two, long rectangular windows for direct observation of the specimen. A work-accelerated annular filament gun moves vertically on a twin-screw traversing mechanism at speeds which can be varied from 0.1 to 30 inches per hour. The vacuum system includes an integral 9-inch main right-angle valve and liquid N_2 cold trap with a 6-inch diffusion pump that is backed by a 30 cfm mechanical pump. A current-regulated 10 kw power supply can be varied from 0 to 30 KV. An auxiliary vacuum-chamber port provides a capability for mounting a Pierce-type self-accelerating gun for remote bombardment.

8) A cold-wall induction vacuum furnace (Figure 6) has been acquired that can provide melting and alloying of ceramics. The stainless steel vacuum chamber (18-inch I. D. and 26 inches long) is evacuated by a 4-inch vacuum system that contains a main valve, a booster pump, and a 15 cfm mechanical pump. A coaxial mechanism provides for both the introduction of power as well as a crucible-tilting capability. Induction power is furnished by the same 30 kw unit mentioned previously under Item 3 above.

9) A high-pressure arc furnace (Figure 7) is being assembled that will be capable of operating at 3000 psi, 1000 amps, and 30 volts. This furnace allows the melting and casting of refractory materials which possess a high vapor pressure to sample sizes of 2-inch diameter by 4 inches long.

10) A high-vacuum evaporator (Figure 8) has been acquired. The vacuum chamber consists of a pyrex bell jar (18-inch diameter and 30 inches long) that can be evacuated to 10^{-6} Torr by a 4-inch vacuum system. The 500-amp power supply provides for resistance heating within the vacuum chamber.

Other miscellaneous processing equipment includes the following:

1) A glo-bar element muffle furnace for general heat treatment applications. The maximum operating temperature is 1350°C, and inside muffle dimensions are 3 inches by 15 inches and 6 inches high.

2) An abrasive machine (Figure 9) operated under high-pressure gas enables the cutting, sectioning, and cleaning of ceramic or metallic specimens.

3) A jar mill apparatus for mixing and grinding.

4) An ultrasonic cleaner.

5) A planetary mixer.

6) A V-blender.

7) A 30-ton hydraulic press for cold pressing.

A specialized machine shop for preparing test specimens to specific sizes or tolerances is also essential to a ceramic materials research program; therefore, the following machine tools have been installed within the Aerospace Ceramic Laboratory area:

1) A Thompson surface grinder

2) A Hardinge horizontal milling machine

3) An Elox electrical discharge machine (10 watt)

4) A Unirad radial grinder

5) A Johansson core-drilling machine (1 HP)

6) Miscellaneous--6-inch bench lathe, 10-inch bench saw, 1/2-inch drill press, 7-inch pedestal grinder, and a Do-All band saw.

B. EXPERIMENTAL RESEARCH

Although the construction of the laboratory has seriously hampered significant research progress, work of a qualitative exploratory nature has been performed. Only upon completion of the construction phase, now estimated at 15 September 1962, can the full resources of personnel and equipment be utilized.

High-purity elemental materials for compound synthesis have been obtained and are tabulated below.

Material	Purity, Percent	Supplier
Columbium	99.8	DuPont Co.
Graphite (Grade AGOT)	99.9 +	National Carbon Company
Titanium	99.9	Titanium Metals Corp. Amer.
Boron (Amorphous)	95-97	U.S. Borax Corporation
Boron (Crystalline)	98.5	U.S. Borax Corporation
Zirconium	99.8	Wah Chang Corporation
Hafnium + (2% Zr)	97.0	Wah Chang Corporation
Tantalum	99.9	Wah Chang Corporation
Tungsten	99.9	Wah Chang Corporation
Molybdenum	99.9	Wah Chang Corporation

For purposes of comparison with properties of the materials that are fabricated from the high-purity elements listed above, certain commercially available carbide and boride powders have been obtained. These powders are listed on the following page.

Powder	Purity, Percent	Mesh Size	Supplier
Boron Carbide (B_4C)	98+	-325	Plasmadyne Corporation
Chromium Carbide (Cr_3C_2)	98+	-150 +325	Plasmadyne Corporation
Tantalum Carbide (TaC)	98+	-200 +325	Plasmadyne Corporation
Titanium Carbide (TiC)	98+	-200 +325	Plasmadyne Corporation
Tungsten Carbide (WC)	98+	-200 +325	Plasmadyne Corporation
Zirconium Carbide (ZrC)	98+	-150 +325	Plasmadyne Corporation
Zirconium Diboride (ZrB_2)	98.4	-150 +325	Plasmadyne Corporation
Tantalum Diboride (TaB_2)	98.2	-200 +325	Plasmadyne Corporation
Titanium Diboride (TiB_2)	98+	-100	American Potash and Chem. Co.

Laboratory work performed to date has been concerned with upgrading lower purity boron to higher levels of purity and development of melting and casting techniques using titanium carbide.

1. <u>Borides</u>

Upon reviewing the commercial market, it was ascertained that boron of a purity equal to or greater than 99.5% was prohibitive in cost at $1,000 per pound. Therefore, experimental work was initiated to determine the feasibility of upgrading lower purity boron (95 to 98.5%) to improved purity levels.

In one of the early experiments a quantity of amorphous boron (95 to 97%) was charged to the arc furnace and its behavior studied during melting. The material fused but excessive splatter occurred which was attributed to absorbed gas, metallic magnesium (present as a major impurity), and poor thermal shock resistance. It was immediately obvious that a pre-treatment is required in order to make the boron react more stably in the arc. A first attempt at successful pre-treatment was an experiment wherein the amorphous boron powder was subjected to a high-temperature vacuum treatment.

A vacuum retort assembly was constructed and fitted to the glo-bar muffle furnace. A small quantity of the amorphous boron powder was charged to the retort and exposed to a temperature of $1000^{\circ}C$ (limitation of the stainless steel retort) for three hours. After cooling, it was observed that the powder had changed color from a definite black to a brownish-black. An examination of the condenser baffles and the vacuum pump oil revealed some contamination by a fine particulate matter of unknown composition. Pressure increases during the heating cycle also indicated that the boron powder liberated gaseous impurities. The powder was then charged to the arc furnace and subjected to the same melting conditions that were present previously. Substantial improvement in the boron's behavior in the arc was noted, but better performance is still desired. It is expected that higher purity can be gained by using the inconel retort, now being constructed, which allows higher distillation temperatures for longer periods of time. It can be concluded from these experiments that a qualitative increase in purity has been obtained for amorphous boron.

In another experiment a sample of the 98.5% crystalline boron was also arc melted with the behavior of this material being much better than that for the amorphous boron of lower purity.

It is of immediate interest to study both materials in the arc-melted condition by metallographic and chemical techniques for comparison purposes.

2. Carbides

Homogeneous bulk titanium carbide from high-purity titanium (sponge) and high-purity graphite (Grade AGOT) was prepared by fusing the elemental materials in the arc furnace repeatedly. Both graphite and copper hearths were used to evaluate the problem of carbon stoichiometric control. After consolidation of melts, the bulk carbide was cast into a right circular cylinder specimen (1/2 inch diameter by 2-1/2 inches long) as shown in Figure 10. Casting of the slug was done in a graphite hearth and mold using a graphite electrode under arc conditions of 3000 amps and 30 volts. The ingot is being evaluated by both metallographic and chemical analyses.

The casting of titanium carbide into this size specimen demonstrates the successful operation of the arc furnace and all applied melting and casting techniques. In processing a material with a melting point of approximately 3000°C, one can conclude that similar techniques should allow the successful casting of all the refractory metal diborides. However, the remaining refractory carbides of Zr, Cb, Hf, and Ta all have higher melting points and it is possible that some unknown difficulties may be experienced in these systems.

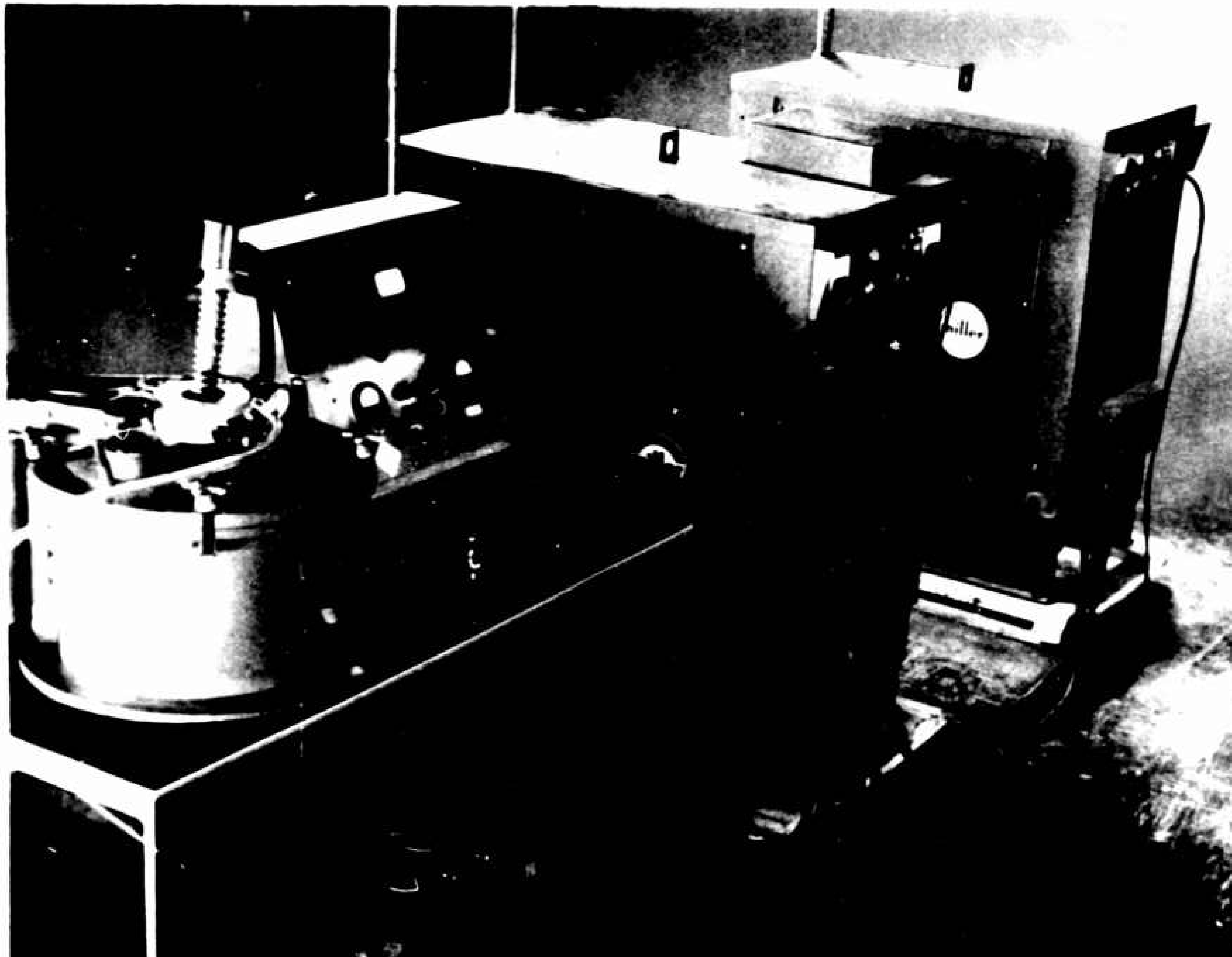

Figure 1. Non-Consumable Electrode Arc Furnace and Power Supply

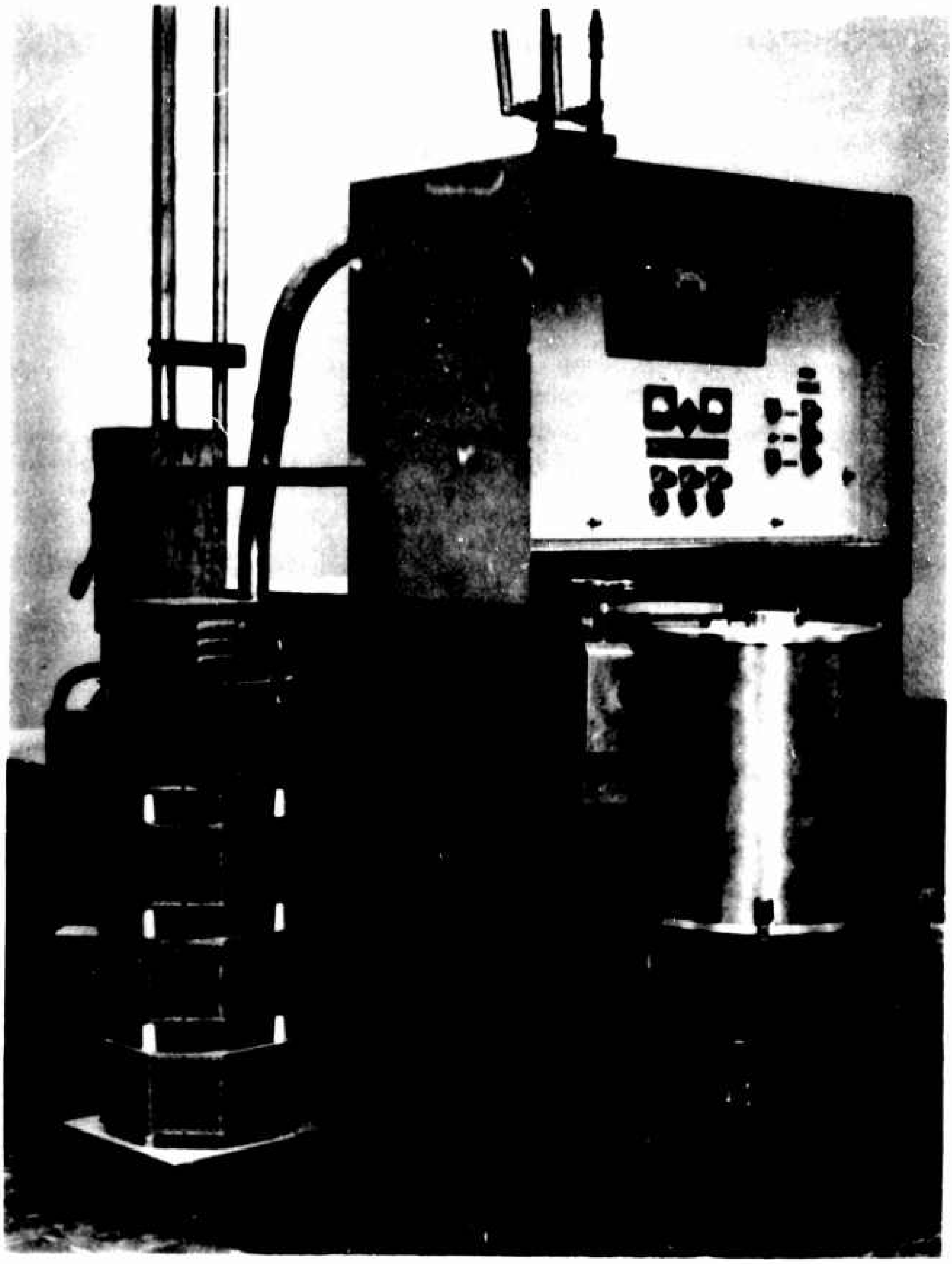

Figure 2. High-Temperature Resistance-Heated Vacuum Furnace

Figure 3. High-Temperature Ho Press

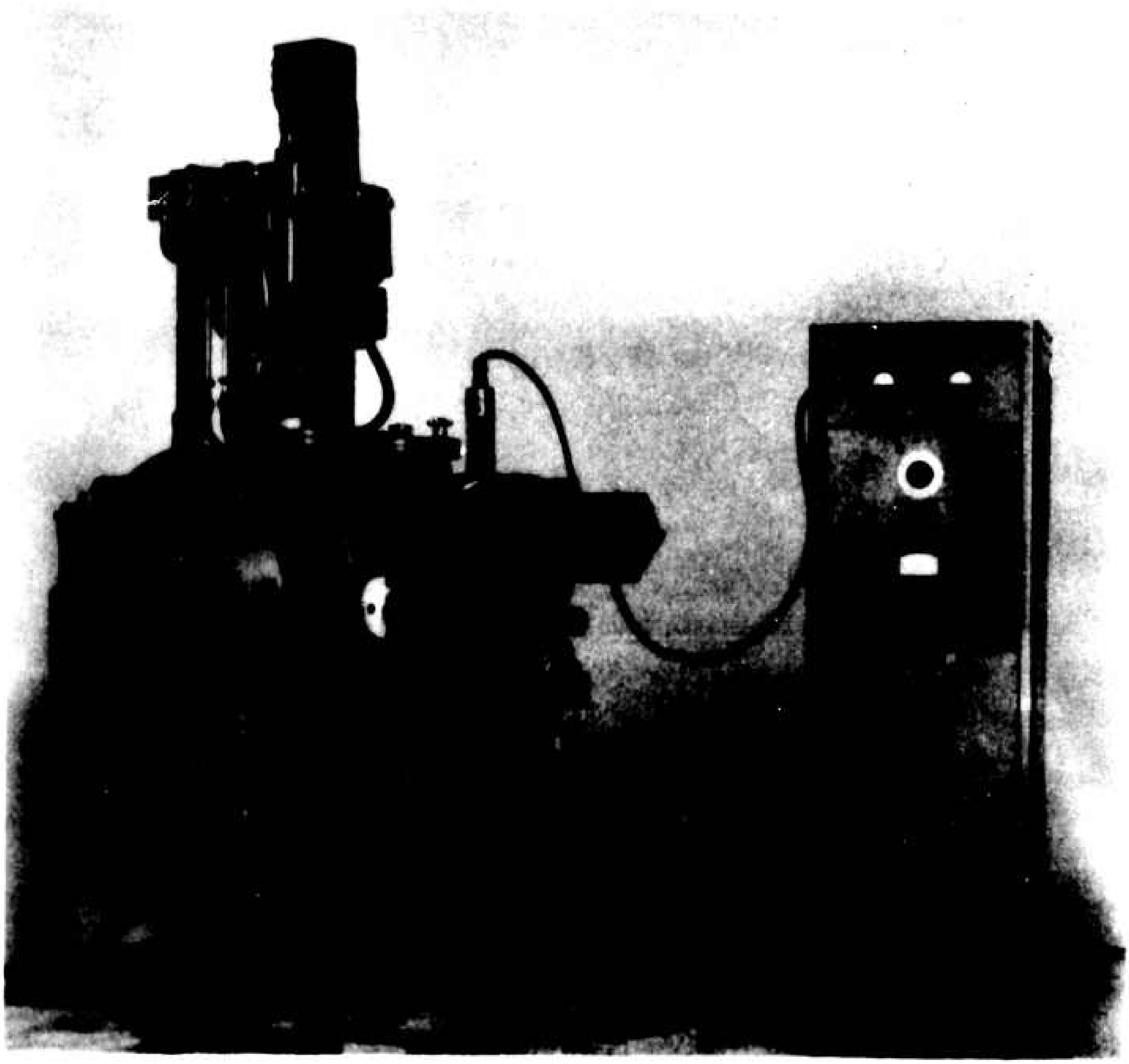

Figure 4. Arc Melting Skull-Casting Vacuum Furnace

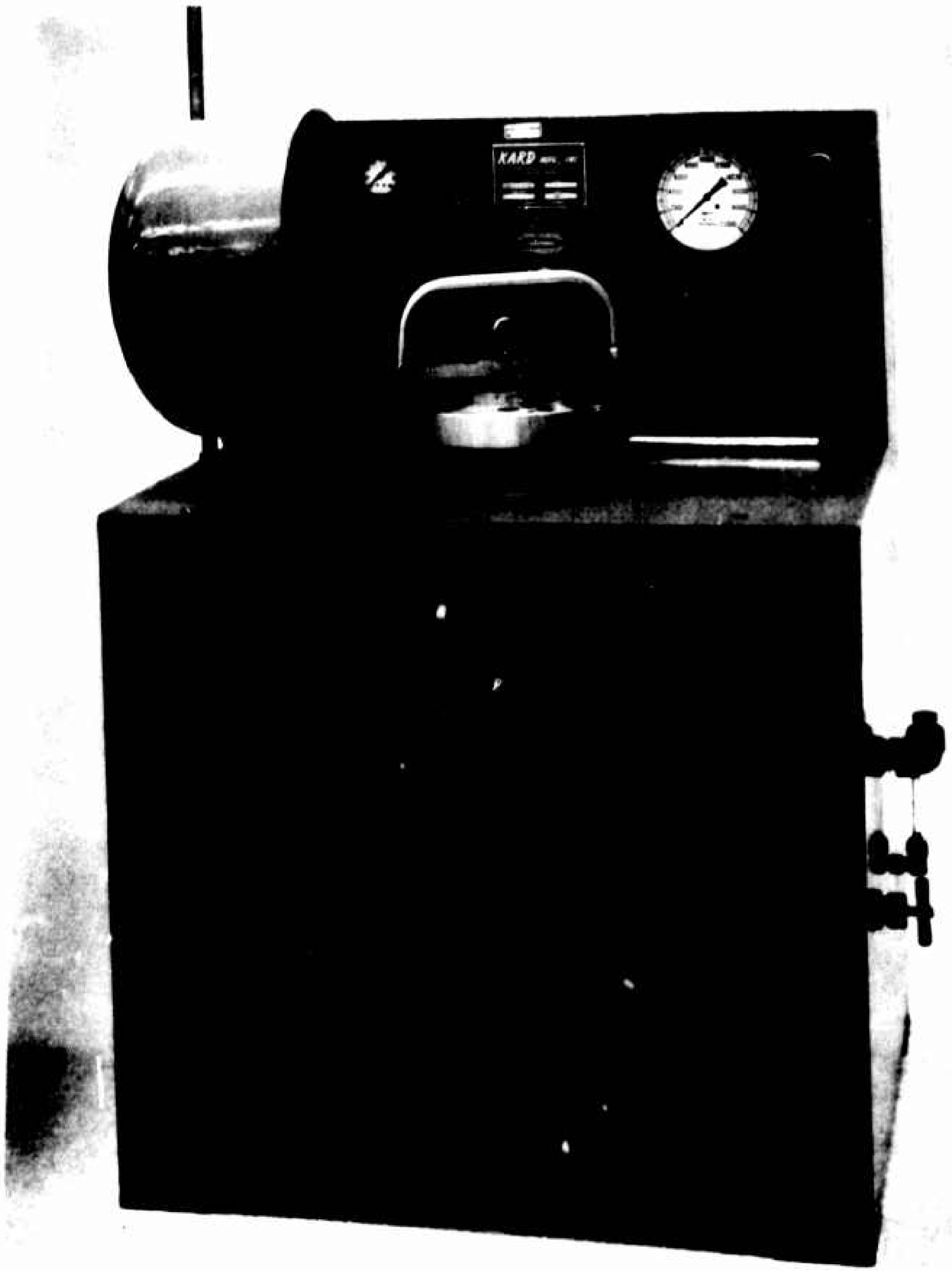

Figure 5. Isostatic Press

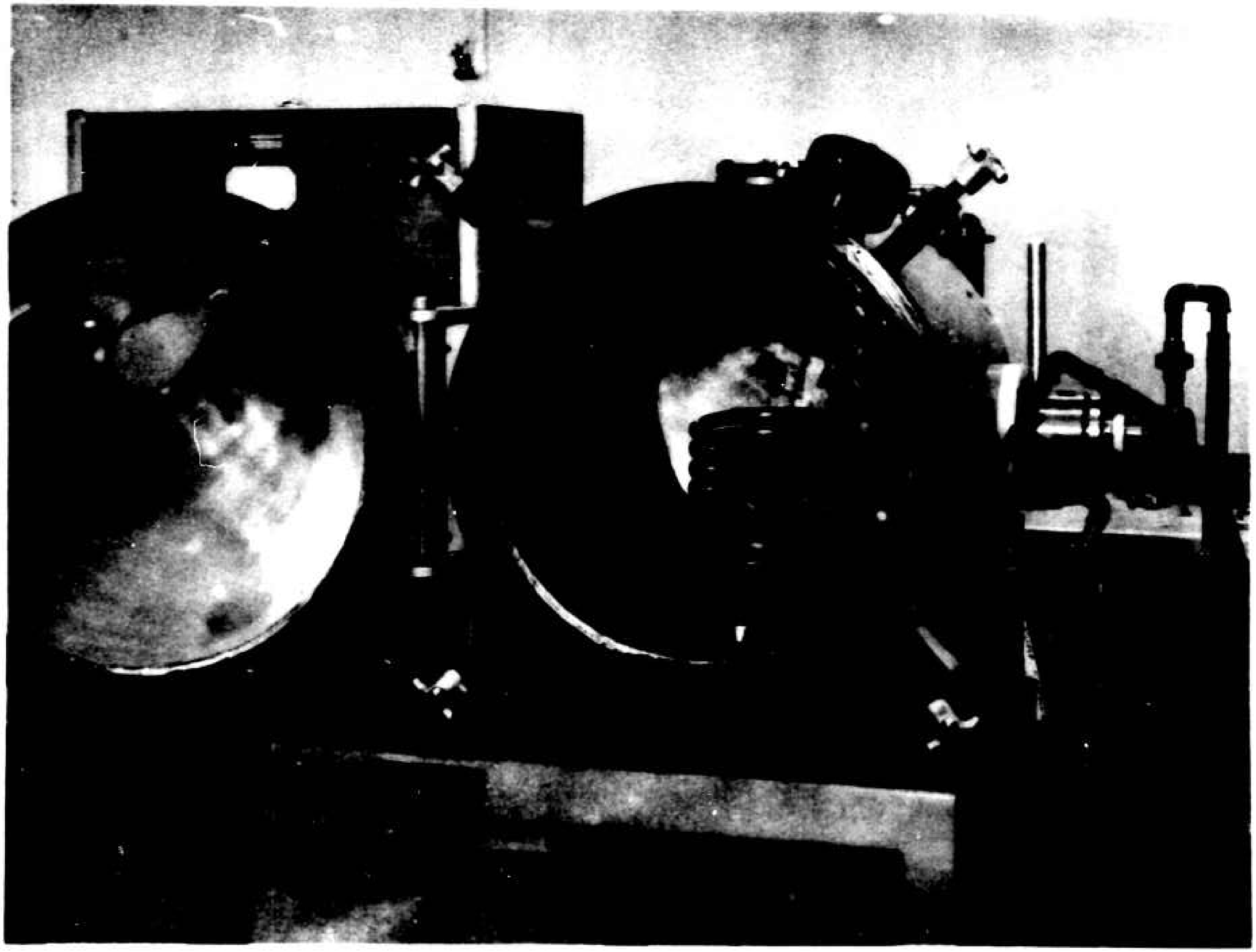

Figure 6. Vacuum Induction Furnace

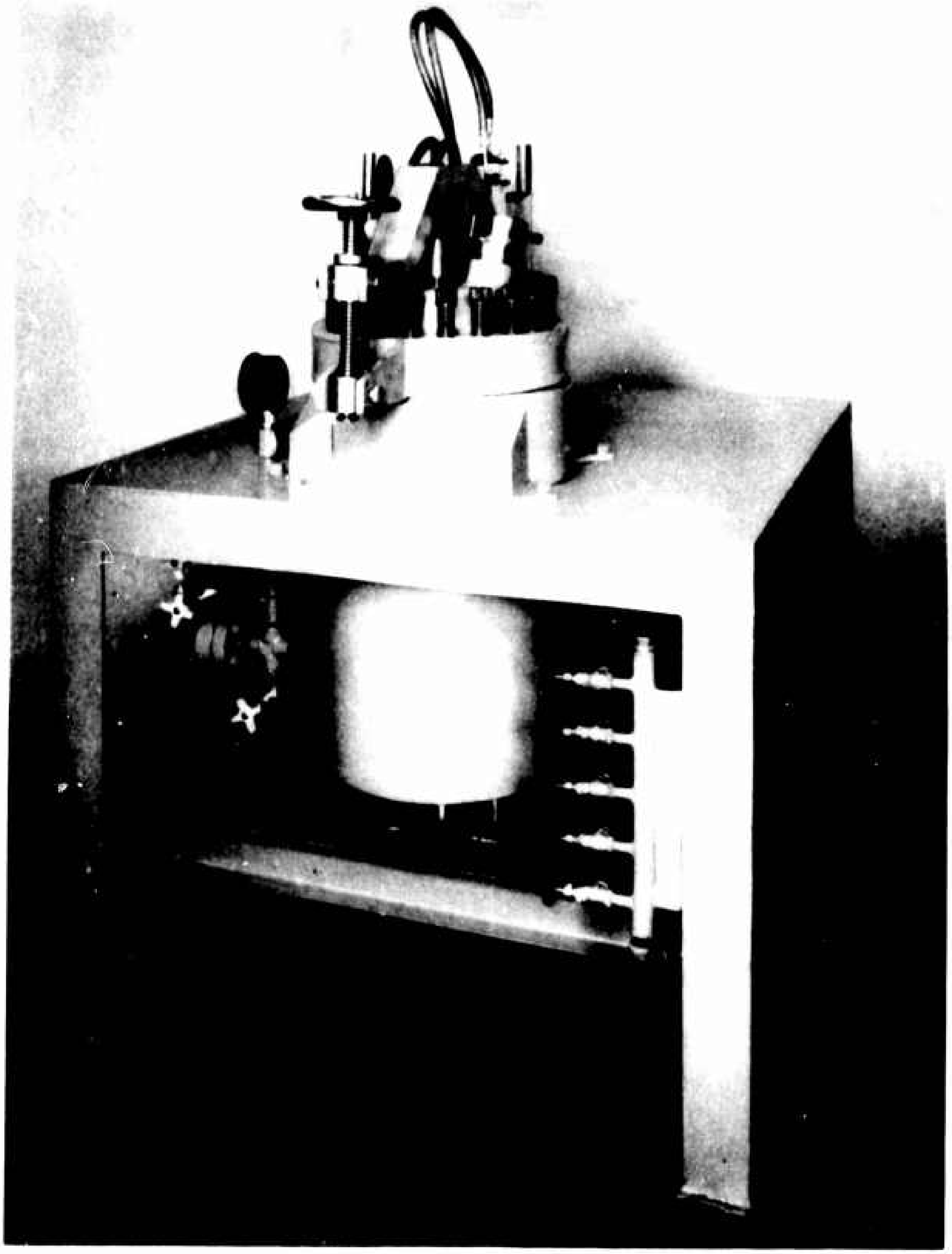

Figure 7. High-Pressure Arc Melting Furnace

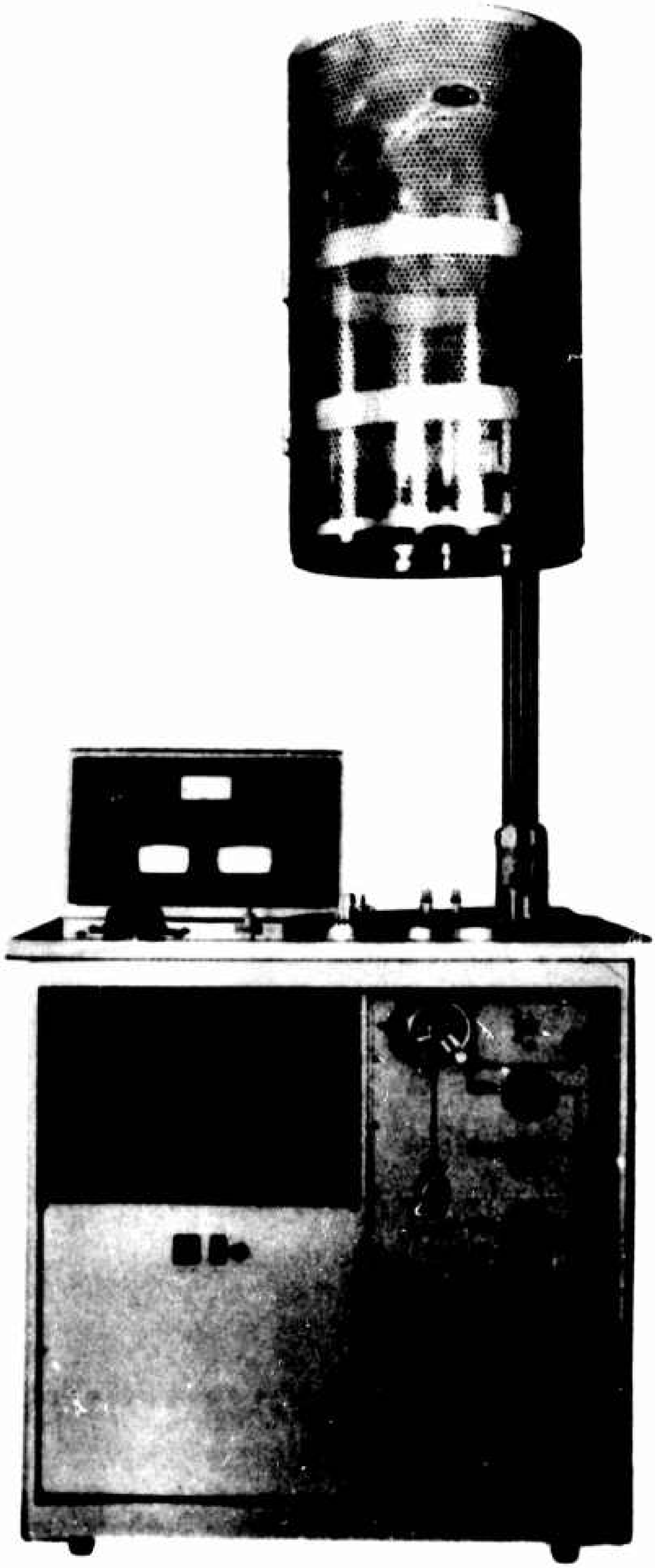

Figure 8. High-Vacuum Evaporator

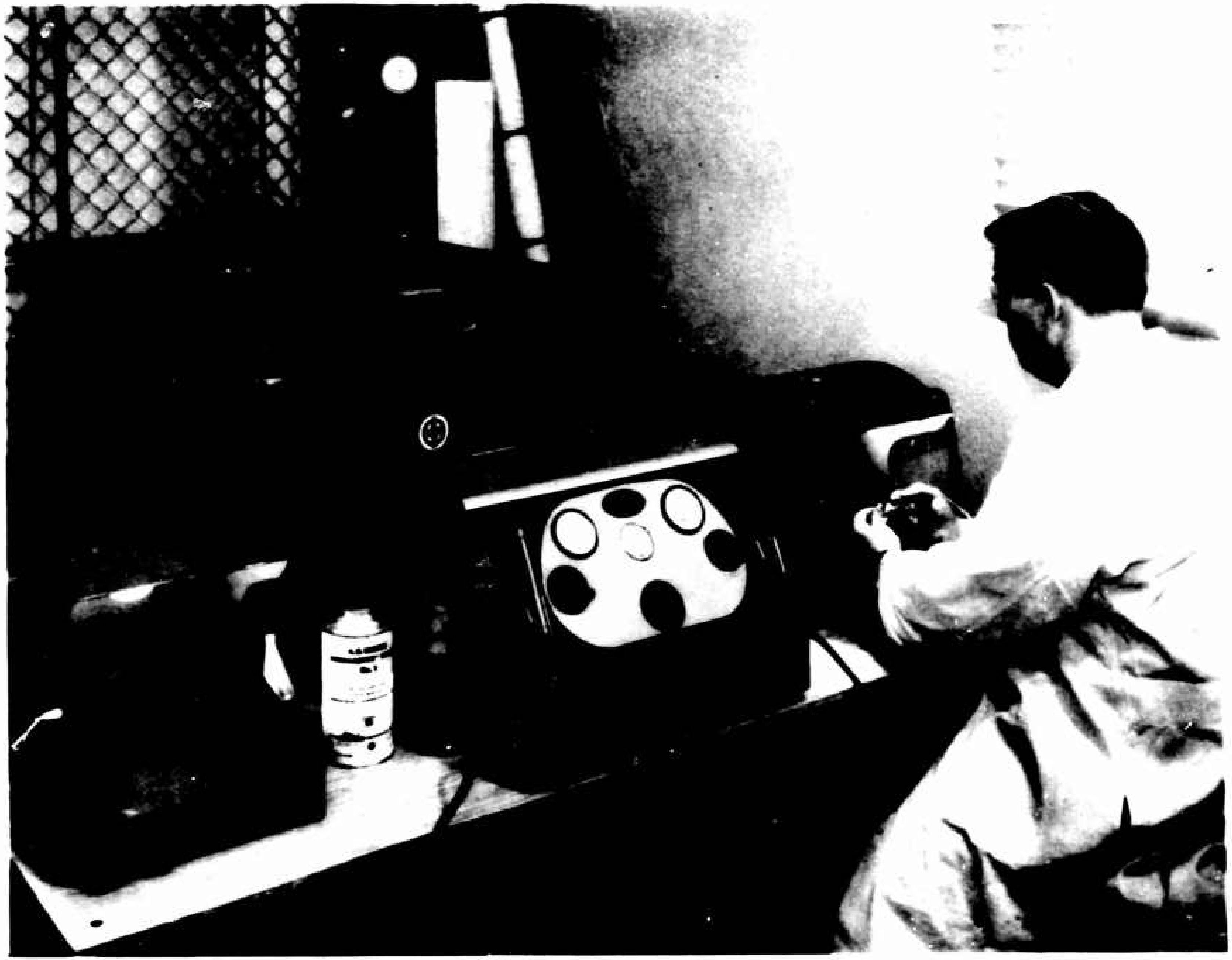

Figure 9. Abrasive Cleaning - Cutting Machine

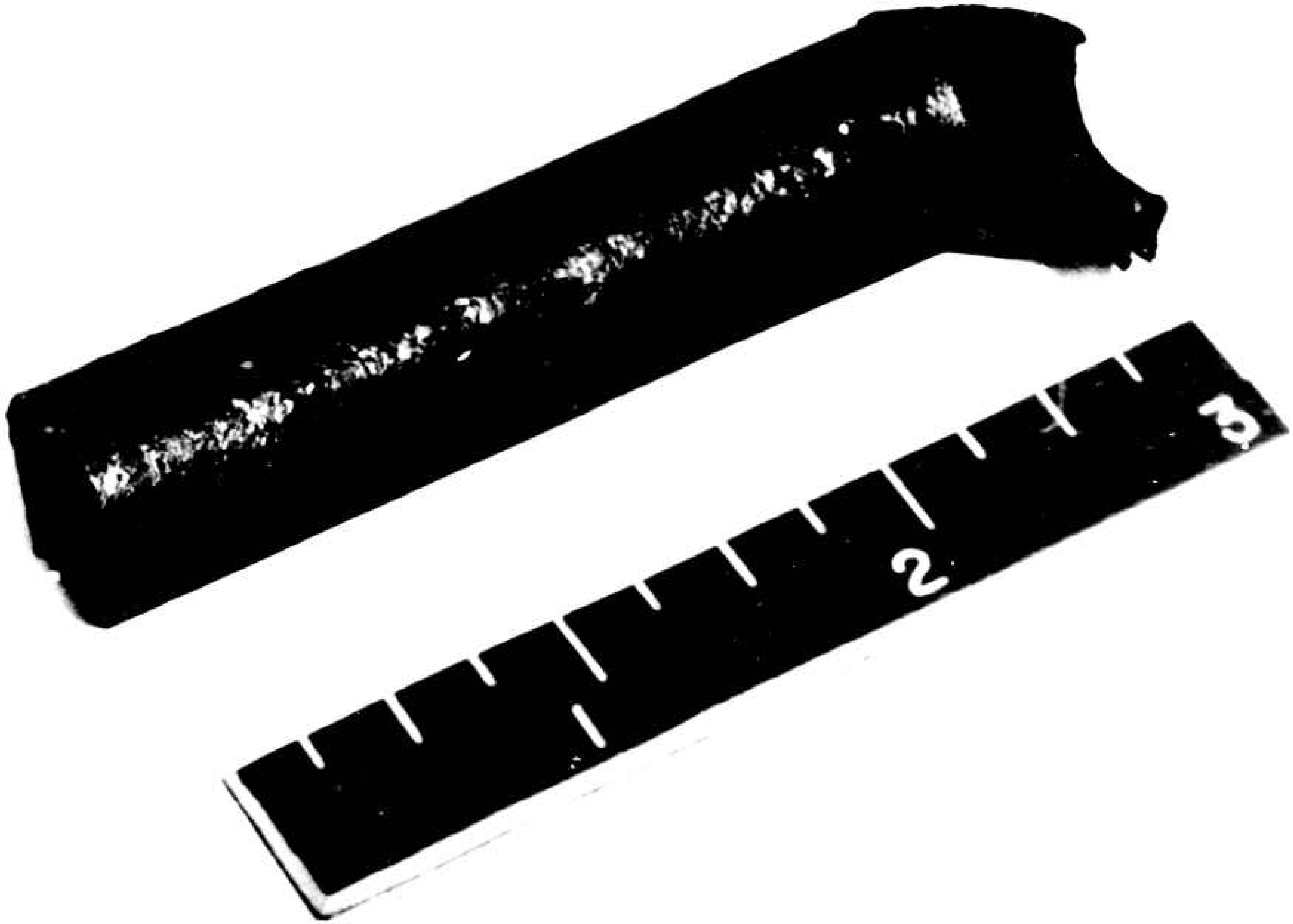

Figure 10. Arc-Cast Titanium Carbide

REFERENCES

1. E. G. Kendall, J. D. McClelland, "Materials and Structures Applied Metallurgical Research Program," Aerospace Corporation, Rpt TDR-930(2240-62)TR-1, 28 February 1962.

2. E. G. Kendall, "Materials and Structures Applied Ceramic Research Program," Aerospace Corporation, Rpt TDR-930(2240-63)TR-1, 28 February 1962.